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GOLD BERYLLIUM ALLOY AND METHOD OF MAKING SAME

Harold Hirsch, Chicago, Ill., and Michael Kowalchik, Rahway, N. J., assignors to the United States of America as represented by the United States Atomic Energy Commission

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2 Claims. (Cl. 75—22)

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This invention relates to a novel composition and the method for making the same and in its more particular phases describes a method for producing ductile, malleable and machinable compositions containing beryllium which retain the increased hardness afforded by addition of beryllium.

A material containing a substantial amount of beryllium but yet very dense and ductile is desirable for use in handling corrosive materials in indicator and other writing points and in particular for use in certain type neutronic reactors. In the past attempts to produce such a material combining density, ductility and malleability have not succeeded.

It has long been known that most alloys containing a major portion of soft dense metals such as gold, lead or the like are relatively soft and as a consequence of limited utility. This fact is particularly true of the binary systems of such metals. U. S. Patent No. 2,015,499 to Smith points out that substantial hardness can be imparted to gold by the addition of up to 5 per cent by weight of beryllium. However, by reason of the methods employed and particularly the formation of the alloy at temperatures in excess of about 500° C., the resultant product is generally brittle or even frangible and is difficult to machine in the as-cast state. See M. Hansen, "Aufbau der Zweistofflegierungen," Edwards Bros. Inc., 1943, page 205. Such a product is further limited in its utility in that it does not have other desirable physical characteristics.

Furthermore, while a number of alloys containing varying percentages of beryllium and gold or other soft matrix metals have been made by casting the metals many disadvantages have been encountered. For example, undesirable impurities such as oxides as well as in homogenieties such as voids are introduced into the alloy which destroys the utility thereof for many purposes. This is particularly true when the neutronic properties of the material are important. While it is true that refinements in the casting process may be employed to obviate foreign inclusions or contamination or even structural defects, such steps are cumbersome and expensive and by no means eliminates the embrittlement which takes place in beryllium containing alloys.

It is well-known that coherent metal bodies may be formed by the application of extremely high compressive pressures to metal powders in the absence of any externally applied heat. In such processes, the pressure alone is relied upon to bond the discrete particles together and if the said pressure is rapidly applied it appears that inter-particle friction is sufficient to raise the local temperature at the interface between

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particles to a point at which such bonding takes place. Such cold pressing methods are generally employed with similar particles and up to the present no such method is known which permits the addition of extraneous elements in particle form to improve the physical properties of the resulting product. On the other hand, in a hot pressing operation, as it is generally understood, the temperature employed is within the range in which diffusion of the materials takes place and a substantial amount of interstitial alloying occurs. With materials such as beryllium, it has now been found that any substantial amounts of diffusion into the soft matrix metals such as gold, lead, or the like will result in embrittlement and the employment of pressure during the heating process will not avoid this result. If no heating is employed in the fabrication of coherent bodies containing such materials, the increase in hardness, density and strength does not come up to desired and predetermined standards.

It is apparent, therefore, that prior to this invention there was no composition known containing beryllium and soft dense matrix metals which was ductile and malleable and which retained the increased hardness afforded by the addition of beryllium.

It is thus seen to be an object of the present invention to provide a novel mixture of gold and beryllium as well as novel steps in the manufacture thereof of such a nature as to define said composition.

It is a further object of the present invention to provide a simple method for preparing mixtures of beryllium and a soft dense matrix metal while avoiding the inclusion of impurities or contaminants.

It is a further object of this invention to provide a method for preparing a dense, hard and yet ductile and malleable continuous uniform metal composition containing beryllium which has very low porosity.

It is another object of this invention to provide a process for preparing a dense metallic composition of beryllium and soft matrix metal or metals which has uniformly reproducible properties.

It is a still further object of this invention to furnish a process for preparing a metallic composition of beryllium and soft matrix metal or metals which is dense but which is not brittle and which thus can be easily machined.

It is a further object of this invention to provide a process for preparing a dense metallic composition of beryllium and soft matrix metal or metals which can be readily plated with other metals such as gold or nickel.

It is a still further object of the present invention to provide a method for preparing bodies containing a mixture of beryllium and soft dense matrix metals, which exhibits more desirable physical properties than were heretofore obtainable.

Another object of the present invention is to provide a method for preparing a composition containing in excess of 5 per cent beryllium and at least one soft matrix metal, which is harder than the matrix metal and which possesses substantially the same characteristics of machinability, ductility and the like of said matrix metals.

The above objects are achieved by preparing an intimate mixture of powders beryllium and at least one soft matrix metal, the said mixture containing a minor proportion of beryllium and a major portion of matrix metal by weight, heating the said mixture to a temperature substanstantially below the sintering temperature and subjecting the mixture, while at an elevated temperature, to a mechanically applied compressive pressure.

While in its elements the said method appears to outline a simple and well-known hot pressing operation, it will be apparent to one skilled in the art that this is not the case. In fact the process as outlined herein, may be termed a "warm pressing" operation because it is carried out below the temperature at which any substantial amount of alloying takes place at the interface of the discrete particles of the elements employed.

The method of this invention produces a dense malleable beryllium composition which does not contain impurities or contaminants and which exhibits many desirable physical properties such as denseness, hardness, ductility, malleability, and low porosity.

One mode of operation in accordance with the present invention may be discussed in connection with the preparation of a product of beryllium and dense soft matrix metal or metals in which the beryllium is employed as a pure metallic powder, for example, of about minus 400 mesh particle size made by methods well-known in the arts. The dense soft matrix metal or metals in like manner are employed in the form of a metallic or powder particle size, for example, of about minus 200 mesh particle size. The powders are then thoroughly mixed in a ball mill. The mixture of beryllium and dense soft matrix metal or metals is incorporated in the die. The powder is lightly compacted and the die heated for a predetermined period of time to a predetermined temperature which is well below the sintering temperautre of the mixture as well as below the temperature at which diffusion takes place or at which a substantial quantity of eutectic is formed. A compressive force is then applied in a manner and to an extent depending upon the composition of the mixture and the nature of the dense soft matrix metal or metals while the composition is held substantially at the predetermined temperature.

In carrying out the method of this invention it is essential that the powders be thoroughly ground and mixed in order to obtain a uniform and homogeneous mixture of materials of such widely different densities.

It is preferable to carefully adjust the degree and duration both of temperatures and compressive force used in order to obtain a product of the necessary physical properties. The final properties of the mixture of beryllium and soft matrix metal depend to a large extent on the temperature to and at which the mixture is heated. With some dense soft matrix materials a narrow temperature range is quite critical. As an example the range of 300° C. to 400° C. is very critical for beryllium-gold mixtures made in accordance with the procedures outlined herein. One reason for this critical limitation is the marked variation of the coefficient of expansion in this range which is probably caused by a permanent phase change. For a mixture consisting of 8.45 per cent beryllium by weight and 91.55 per cent gold by weight the coefficient of expansion is linear up to 300° C., but in the range from 300° C. to 400° C. the change in dimension is very large and accounts for most of the length change of 8.2 per cent. Thus an undesirable permanent volume change occurs when the mixture is heated for a period of time at 400° C. and lower densities result.

On the other hand a minimum temperature is desirable to permit plastic flow of the matrix metal. This temperature is of course predetermined and related to the amount and method of applying the compressive force. For example, as will be pointed out in greater detail later, ten tons per square inch is the preferred minimum compressive force for preparing the gold-beryllium mechanical mixture at which pressure a temperature of 400° C. is required for the production of a finished coherent brick having desirable characteristics. When all these factors are taken into consideration, it is found that 300° C. to 350° C. is the preferred temperature range for preparing a desirable gold-beryllium mechanical mixture comprised of, for example, 8.45 per cent beryllium by weight and 91.55 per cent gold by weight and the pressure applied with this preferred temperature is about 50 tons per square inch. The desirable temperature range, naturally, varies with change in composition. When the mixture is compressed at a temperature lower than the predetermined minimum value the composition tends to laminate. On the other hand when the temperature is permitted to rise appreciably above the predetermined maximum the resultant product becomes very hard and somewhat similar to the alloys containing beryllium produced by casting. This is true because at temperatures of about 500° C. and above diffusion between beryllium and most matrix metals takes place, and a substantial amount of brittle eutectic is formed. For example, if a mixture containing beryllium is pressed at room temperature and then sintered in either an atmosphere of hydrogen, air or in vacuo, a very frangible product results which possesses a laminated structure and which cracks in a direction transverse to the pressure axis.

It is also preferable to control the length of time during which the mixture is heated in order to insure that all parts of the mixture are brought to a uniform temperature and thus to insure that a continuous uniform metal composition is produced. Generally this heating takes place before the application of substantial compressive force, although in some cases this is not necessary.

The said force may be applied in full and continuously during heating, or slowly increased as the heating progresses increasing gradually to a maximum; or it may be applied for a definite period of time after the mixture has been heated to the desired temperature; or it may be applied in as rapid a manner as feasible during or after heating. When the compressive force is applied after the mixture has been heated, it is applied when the mixture is still at the desired pressing temperature. Experience has indicated that to produce a desirable beryllium-gold coherent composition a minimum compressive force of at least one ton per square inch and preferably at least ten tons per square inch is necessary.

As an example of the effect of temperature and pressure on the hardness, density and machinability of a product made by the method of this invention, the results obtained on a number of successive runs under predetermined controlled conditions in the same equipment are listed below. These controlled conditions comprise intimately mixing 8.45 per cent by weight of 200 mesh particle size beryllium with 91.55 per cent by weight of 170 mesh particle size gold in a standard ball mill, placing enough of said material to make a rod one-half inch long if calculated density is obtained in a hardened steel cylindrical compression die provided with two movable opposing plungers one-half inch in diameter and heating said mixture to the temperature indicated in the chart below, immediately removing said heated mixture from the furnace as soon as the said temperature is obtained, and subjecting the same to the compressive force indicated for five minutes and thereafter permitting the mixture to cool in air. The data obtained is as follows:

CHART I

Data indicating effect of temperature and pressure on physical properties of a mixture made according to the method of this invention

Run No.	Temp., °C.	Pressure, tons per sq. in.	Density, grams per ml.	Density, per cent of theory	Hardness Rockwell Scale "B"	Machinability
1	200	55	10.17	94.7	69	Satisfactory.
2	200	70	10.37	96.5	74	Good.
3	250	55	10.38	96.5	75	Do.
4	250	70	10.41	96.8	76	Superior.

It may be seen from these results that whereas under the same temperature conditions an increase in pressure from 55 to 70 tons per square inch improves the density, hardness and machinability, and whereas these same factors are improved by increase in temperature under the same pressure conditions, the best results are obtained by an increase of both temperature and pressure. The temperature must of course be maintained below the value at which substantial diffusion of the metals into each other takes place with the formation of sufficient eutectic to cause brittleness.

Other variations (heretofore noted) of the method of the invention than those listed in the table may of course be used for preparing beryllium-gold compositions comprising 8.45 per cent beryllium and 91.55 per cent gold or for other proportions comprising the same two metals, as will be shown hereinafter in greater detail. In other words the method of the invention in its broadest consideration comprises preparing coherent metallic bodies containing beryllium and at least one dense soft matrix metal. The preferred embodiment of the method of the invention together with two illustrative variations thereof are presented in the following examples for purposes of illustration but they are not intended to be limiting on the spirit or scope thereof.

EXAMPLE I

Beryllium metal powder of minus 200 mesh particle size is prepared by methods well-known in the art. Lead metal powder of minus 200 mesh particle size is prepared by methods also well-known in the art. Predetermined quantities of the said powders are then thoroughly mixed in a standard ball mill. The said mixture is then placed in a compression die of standard design with two movable plungers. A slight, initial compressive force is applied to force said plungers into the die far enough to assure sufficient bearing surface so that when a substantial compression force is applied, the plungers will move freely and will not bind. The said mixture is then hot pressed in the die at a temperature depending upon the mixture and the pressure, for example, 250° C. while a compressive force of 10 tons per square inch is applied, the duration of the operation being five minutes. The temperature is maintained by suitable means such as a resistance furnace element. The compressive force is delivered by standard equipment such as between the platens of a hydraulic press. The resultant product is removed from the die immediately after the completion of the pressing step and is permitted to cool in air.

A mixture of 14.85 per cent by weight beryllium and 85.15 per cent lead when treated in this manner forms a product having a density of about 6.4 grams per milliliter which is practically calculated density. The coherent metallic composition that results has very desirable properties because it is not brittle or porous and is homogeneous and easy to machine.

EXAMPLE II

Beryllium powder of minus 200 mesh particle size, gold powder of minus 170 mesh particle size and lead powder of minus 200 mesh particle size are prepared by methods well-known in the arts. The said metal powders are thoroughly mixed in a standard ball mill.

The said mixture is then compacted into a compression die of standard design and pressed at 300° C. at 50 tons per square inch. The resultant product is then immediately removed from the die.

A metallic composition of the weight ratio 8.38 per cent beryllium, 27.1 per cent lead and 64.52 per cent gold prepared in this manner has a density of substantially 9.7 grams per milliliter which is better than 99 per cent of the calculated value. The hardness is 87.2 on the Rockwell "B" scale. The product is dense, homogeneous and easily machined and is not brittle.

EXAMPLE III

In the presently preferred embodiment of the invention, pure beryllium metal is prepared as minus 400 mesh particle size powder. Pure gold is likewise prepared as minus 170 mesh particle size powder. The two are thoroughly mixed in a standard ball mill. The various parts of the compression die of a standard design are coated with colloidal graphite or aquadag to permit freedom from binding and easy ejection. The mixture of gold and beryllium is then incorporated in the die and the movable portions thereof adjusted. A slight compressive force is then applied to said movable portions. The temperature of the entire assembly is raised to about 350° C. by suitable means, e. g., a resistance furnace element, and this temperature held for one hour. As soon as the die is removed from the furnace it is placed in the press and a compressive force of 55 tons per square inch is applied for 5 minutes during which time the temperature remains above 300° C. The compressive force is released and the resultant product is immediately removed from the die and permitted to cool in air.

When a mixture comprising 8.45 per cent beryllium and 91.55 per cent gold by weight is used, this treatment gives a mechanical mixture of density equal to about 10.6 grams per milliliter which is better than 98 per cent of the calculated density. The hardness of the continuous uniform metal composition is 95 on the Rockwell "B" scale. The product can be machined and is not brittle. It is very uniform and it is not porous. The product has excellent corrosion resistance properties and may be heated to 300° C. without change. It may be plated easily with metals such as gold or nickel either by electrodeposition or decomposition of metal complexes such as carbonyls.

It may be seen that thus contrary to other practices in hot-pressing of powdered metals or casting of beryllium containing mixtures, extremely low temperatures are employed to produce added desirable properties in certain metals and compositions containing them, thus largely avoiding defects inherent in similar compositions prepared by such other methods.

The coherent metallic body that results is very desirable for use, for example, in certain neutronic reactors since it is dense, uniform, hard, malleable and ductile and since it is substantially free of extraneous elements which would absorb neutrons to a material extent.

However, the product is not limited to use in neutronic systems. For example, by reason of the corrosion resistance, taken together with the hardness and malleability not obtainable with gold or similar dense malleable metals alone, it is feasible to use the gold-beryllium products prepared by the method of this invention in electrical devices, in equipment for handling corrosive materials, in indicator and other writing points, and the like.

It must be understood that although the methods of the examples have been stated with the conditions given very specifically, the conditions may be varied. The temperatures and pressures may be varied as indicated hereinabove. Although definite particle sizes have been stated any particle size or sizes may be used as long as an intimate and homogeneous mixture of the metals results. It is also possible to plate the soft matrix metal on the beryllium and then compact the powders. This is particularly feasible with gold. In like manner although the methods of the examples state definite proportions of the compositions comprising beryllium and lead; beryllium, lead and gold; and beryllium and gold; the method may also be applied to other proportions of these metals or to any mixture comprised of a minor proportion of beryllium and a major proportion of any relatively soft dense matrix metal or metals by weight. These matrix metals may be any soft metal with a specific gravity greater than 11.0 such as lead, gold, thallium, thorium, platinum, or palladium and are commonly known as soft, dense matrix metals.

Such mixtures treated as herein disclosed result in compositions which are decidedly harder than the matrix metal without the sacrifice of ductility, malleability, machinability, purity or homogeniety. These products which result from the method of this invention are coherent metallic bodies which are not alloys of the metal but are instead mechanical mixtures which may or may not contain some but not substantial alloying of the metals at the interfaces of the beryllium and the dense matrix metal or metals. The product is a continuous uniform metal composition. The resulting compositions cannot be made by any other method and possess properties wholly differing from similar compositions prepared by other methods such as straight alloying. As a result this product can only be defined as the product of the processes of the invention.

It is to be understood that when the product is said to be a continuous uniform metal composition by this is meant that finite cross-sections of the metal have substantially similar physical properties such as density, hardness, ductility and malleability.

The foregoing examples are given for the purpose of illustrating the present invention but are not intended to be limiting on the scope thereof. As many widely different embodiments of the invention can be made without departing from the spirit and scope thereof, it is to be understood that this invention is not to be limited except as indicated in the appended claims.

What is claimed is:

1. A process for preparing coherent metallic bodies of about 8 per cent by weight beryllium and about 92 per cent by weight gold, which comprises intimately mixing powders of said elements in the aforesaid proportions, compacting the mixture in a die, heating the powders to a temperature of about 350° C. for about one hour and then compressing the mixture with a compressive force of at least 50 tons per square inch for five minutes.

2. The product of the process of claim 1.

HAROLD HIRSCH.
MICHAEL KOWALCHIK.

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